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(54) A METHOD OF FORMING A LAMINATE

(71) I, OLE-BENDT RASMUSSEN, a Danish subject, of 7 Topstykket, DK-3460 Birkerod, Denmark, do hereby declare the invention, for which I pray that a patent may be granted to me, and the method by which it is to be performed, to be particularly described in and by the following statement:—

Various ways are known of bonding films together to form a laminate. If the films are made by extrusion it is common to extrude also on one or both of the facing surfaces of the films an adhesive polymer such that mere pressure between the films, often combined with the application of heat, results in bonding.

A method acording to the invention of forming a laminate comprises orienting by stretching two or more films while they are pressed together, such that the surfaces of the films are in contact, the surfaces of the films being of polymeric material such that, in the absence of the stretching, the surfaces of the films do not bond to one another but, as a result of the stretching, they do bond together. This process can be carried out at room temperature.

The invention also includes apparatus for carrying out this method. This apparatus comprises means for advancing two or more films towards a transverse stretching system, and means for superposing the films in the system, in which the transverse stretching system includes means for applying pressure along lines extending substantially in the longitudinal direction of the films and for simultaneously stretching the films in the transverse direction, thereby forming a laminate having a configuration of temporarily substantially evenly distributed substantially longitudinal pleats.

Thus the stretching is preferably conducted continuously transversely on continuous lengths of films, to form a laminate having a configuration of temporarily substantially evenly distributed substantially longitudinal pleats, by applying pressure along lines extending substantially in the longitudinal direction of the films to stretch the films in the transverse direction.

This is preferably achieved by passing two or more films through the nips of two or more pairs of intermeshing grooved rollers in which the grooves extend substantially in the longitudinal direction of the sheet. Thus pleats are formed during the transverse stretching, and bonding occurs. Preferably the pleats formed by passage through one said pair are flattened out before passage through the next said pair.

One at least of the grooved rollers used may be heated to improve bonding. Often, however as described above, all the stretching under pressure is conducted substantially at room temperature.

The method of the invention is preferably combined with one or more longitudinal stretching steps, the resultant laminate thus being biaxially stretched. The longitudinal stretching should preferably take place after the transverse stretching is essentially finished.

The purpose of carrying out the longitudinal stretching subsequent to the transverse stretching is to avoid an exaggerated longitudinal striation of the film which may be formed when the grooved rollers or similar devices work on longitudinally oriented film.

When specifically high tear propagation and puncture resistance is desirable, and relatively low yield point is allowable, the stretching is preferably carried out on machinery which allows a substantial transverse contraction. There is hereby obtained a higher elongation to break. In order to allow the contraction at the same time as the longitudinal stretching is carried out—as is usually desirable—in a narrow zone, the laminate is preferably supplied with very fine, longitudinal pleats—in analogy to what is disclosed in U.S. Patent No. 3,233,029. In this connection, a suitable result is usually obtained when the fine pleats formed by the last step of the lateral stretching are maintained in the sheet when the latter is fed into the longitudinal stretching zone.

It is normally preferred that at least one facing surface of the films that are to be

welded should comprise a layer of a polymer having a lower melting point than the remainder of the film. Both facing surfaces may comprise such a layer. The layer may be in the form of stripes or spots of the lower-melting polymer. One facing surface may comprise a lower-melting polymer while the other may comprise a layer of spots or stripes of a release polymer, that is to say one that reduces the strength of bonding.

The films are preferably made by extrusion of molten polymeric material and most preferably by extrusion of a blend of polymers which are incompatible such that on solidification the blend comprises a dispersion of one polymer in a matrix. Preferably each extruded layer of polymeric blend is attenuated while molten before, during or after extrusion to distort the polymer in each layer into a fibrillar grain structure having a predominant orientation of splittability after solidification.

Preferred methods of conducting this extrusion, together with a description of final products made by the method, and full description of materials that can be used as the polymeric blend and as the lower-melting polymer, are given in my copending application 29807/74 (Serial No. 1,526,722) from which the present application is divided.

The invention is illustrated in the accompanying drawings in which:

Fig. 1 is a process-line showing a method.

Fig. 2 is a detail of the "grooved rollers" which perform the transverse stretching in uneven zones, called "striations".

Fig. 3 is a schematical sketch, on an enlarged scale, of the pattern of the striations and the orientation therein, of a film cross-stretched according to the process-line of Fig. 1.

Fig. 4 is an enlarged cross-section of the film of Fig. 3 as actually observed by microscopy. However, for the sake of clarity, the thickness is shown on a scale twice that of the width.

Referring to Fig. 1, section Q is the part of the line where cross-stretching and lamination is conducted on sheet 79 and section R is the longitudinal stretching line. Sheet 79 comprises a pair of films that have been supplied separately and brought together but not finally bonded into a laminate prior to the first rolls 72.

The system of rollers in section Q consists of driven nip-rollers (71), driven grooved rollers (72), idle rollers (73) and rollers (74) having a cross section resembling a banana. The banana rollers (74) after each step serve to draw out the pleats produced by the transverse stretching. Over the idle roller (75), the sheet (79) enters section R, the longitudinal stretching line, where it is drawn through a water bath (76) serving to remove heat generated by the stretching and maintain a suitable stretching temperature, e.g. at 20°—40°C. Finally it is wound onto a bobbin (77).

The arrow (78) indicates the machine direction.

In Fig. 2 a pair of driven grooved rollers (72) are shown in detail with the sheet (79) pressed and stretched between the teeth (80) of the rollers (72).

In Fig. 3 the relative lengths of the arrows in the striations I and II of the sheet (79) indicate the relative amounts of orientation achieved by the biaxial stretching method shown in Fig. 1 and Fig. 2.

In Fig. 3, as well as in Fig. 4, the numbers I and II indicate the striations A and B which generally have varying width and uneven character. Furthermore, it should be noted that the outer layers (81) and (82) of the sheet (79) are not always symmetrical with respect to the thin mid-layer (83). This asymmetry further serves to make a tear fork.

The following are some examples of the invention, in which all percentages are by weight.

EXAMPLE 1

A three-layer tubular film is extruded having the following composition; Middle layer (70% of total): Blend of 85% isotactic polypropylene of gas-phase type ("Novolen") with high atactic content with 15% ethylene-vinyl acetate copolymer (16% vinyl acetate). Both surface layers (one 10% of total, the other one 20% of total): Ethylene-vinyl acetate copolymer (16% vinyl acetate)—to serve as lower-melting layers.

The polypropylene has a melt index 0.3—0.6 according to ASTM D 12 38 condition L, while the ethylene-vinyl acetate copolymer has a melt index 2.5 according to the same ASTM but condition E. The tubular film is extruded from a 1 mm wide slot at 180°C—230°C and drawn to 0.130 mm in molten state. The blow ratio is kept very low, viz. 1.2:1.

Thereafter it is cut helically to a flat film with 45° angle of grain. Two such helically cut films, with their grain perpendicular to each other, and the thinner surface layers facing each other, are fed together at 20°C through 7 sets of grooved rollers— see Fig. 1 and 2. The width of each groove is 1 mm and the width of each ridge is 0.5 mm. The intermeshing of the ridges with each other (difference of level between the tops) is 2 mm. Between each passage through a set of grooved rollers the pleats formed in the laminate are straightened out. By the mechanical work between the grooved rollers, and owing to the presence of the lower-melting copolymer layers, the two films are cold-welded together with relatively low bonding strength,—peel strength measured to 10 gr per cm—and are at the same time cross-drawn.

After the 7 passages at 20°C the laminate

is passed once through a similar set of grooved rollers with the same dimensions and intermeshing, but heated to 120°C, whereby lines of strong bonding are formed. Finally the laminate is longitudinally oriented by drawing in three steps with about 1 cm stretching-gab (so as to minimise the cross-contraction). The last stretching is so adjusted that the total lateral cold-stretch-ratio and the total longitudinal cold-stretch-ratio are equal. The area-stretch-ratio is 2.4:1.

Test results on the product, compared to a heavy-duty bag quality low-density polyethylene film of 85% higher sq.m. weight and melt index 0.3, according to same ASTM condition E. Gauge, show 100 gr per sq.m. for the laminate and 185 gr per sq.m. for the polyethylene film.

Impact strength (measured by falling-ball of diameter: 61 mm, weight 320 gr) for the laminated film of 100 gr per sq.m.: 5:5 m. For the polyethylene film of 180 gr per sq.m.: 2.0 m. Tongue tear resistance: (tearing at velocity 100 mm per min, total specimen width 5 cm, incision length 10 cm). For laminated film of 100 gr per sq.m.: 5.9 kg in the machine direction and 6.8 kg in the transverse direction.

For the polyethylene film of 180 gr per sq.m.: 1.3 kgr. Elmendorf tear resistance (shock-tearing): (this test is a modification of standards aiming at a more symmetrical tearing). Results: For the laminated film of 100 gr per sq.m.: in longitudinal direction 441 kg cm per sq.cm, in cross-direction 344 kg cm per sq.cm. For the polypropylene film of 180 gr per sq.m.: in longitudinal direction 167 kg per sq.cm, in cross-direction 172 kg cm per sq.cm. When a piece of the sheet is delaminated by peeling and the structure is examined by microscope the main layers are seen to have a pronounced fibrous morphology with zig-zagging grain-directions.

EXAMPLE 2

The procedure of example 1 is repeated with the following modifications: The three-layer coextruded film had the following composition: Middle layer (70% of total): Blend of 85% iso tactic polypropylene (same type as in Example 1) with 15% ethylene - propylene - rubber (ab. same melt index as the polypropylene). Both surface layers (each 15% of total) ethylene-vinyl acetate copolymer (same type as in Example 1). The film was more strongly melt-attenuated after the exit from the die, namely by drawing from 1 mm thickness to 0.065 mm (60 gr. per sq.m). Examinations in polarized light showed that the melt-orientation produced corresponded to about 35% uniaxial cold-drawing. After the spiral-cutting, of this film two were bonded with a third film to produce a 3-ply laminate. The third layer, which was placed in the middle, was with longitudinal grain direction obtained by longitudinal cutting of the same film. The lamination and drawing took place on the machinery of Example 1 but all steps were carried out at 20°C, and the apparatus was adjusted to produce a total area-stretch ratio of 2.5:1 by which the final laminate thickness became 72 gr per sq.m. The peel strength of the bonding between the layers was measured to 10 gr per cm. Examination by microscope showed a similar structure as in Example 1. The following test results were obtained:

		Present film 3 layers 72 gr/sq.m.	Comparison un-oriented LDPE—film 184 gr/sq.m.
Impact Strength	gr	1000 gr×m	530 gr×m
Tear propagation strength	MD+)	848 gr×m	307 gr×m
(slow tear)	TD++)	1120 gr×m	620 gr×m
Load at break	MD	11.1 kg	10.6 kg
(1" wide samples)	TD	8.3 kg	10.7 kg
Stretch at break	MD	286%	467%
	TD	347%	620%
Shrinkage			
1 min. 130°C	MD	28%	—
	TD	14%	—
1 min. 155°C	MD	58%	—
	TD	41%	—

+) Machine Direction
++) Transverse Direction

WHAT I CLAIM IS:—

1. A method of forming a laminate which comprises orienting by stretching two or more films while they are pressed together such that the surfaces of the films are in contact, in which the surfaces of the films are of polymeric material such that, in the absence of the stretching, the films do not bond to one another but, as a result of the stretching, they do bond together.

2. A method according to claim 1 in which the stretching is conducted substantially at room temperature.

3. A method according to claim 1 in which the films are pressed together along lines extending substantially in the longitudinal direction of the films and simultaneously stretched in the transverse direction, thereby forming a laminate having a configuration of temporarily substantially evenly distributed substantially longitudinal pleats.

4. A method according to claim 3 conducted by passing the two or more films through the nip of intermeshing grooved rollers in which the groves extend substantially in the longitudinal direction of the film.

5. A method according to claim 4 in which the distance from the middle of one groove to the middle of an adjacent groove on the same roller is substantially 1.5 mm. and the depth of intermeshing is substantially 2 mm.

6. A method according to Claim 4 or Claim 5 in which there is a plurality of pairs of intermeshing grooved rollers and the films are passed in series through the nip of each pair.

7. A method according to claim 6 in which the pleats formed by passage through one said pair are flattened out before passage through the next said pair.

8. A method according to any of claims 4 to 7 in which at least one of the rollers is heated to improve bonding.

9. A method according to any of claims 3 to 7 in which the stretching under pressure is conducted substantially at room temperature.

10. A method according to any of claims 3 to 9 in which the laminate is subsequently stretched longitudinally in one or more separate steps.

11. A method according to claim 10 in which the transverse stretching is essentially finished before the start of the longitudinal stretching.

12. A method according to claim 10 or 11 in which a substantial transverse contraction is allowed during the longitudinal stretching.

13. A method according to any preceding claim in which at least one of the facing surfaces of the films which are to be bonded comprises a layer of polymer having a lower melting point than the remainder of that film.

14. A method according to claim 12 in which the layer of low-melting polymer is in the form of stripes or spots.

15. A method according to any preceding claim in which each film is formed by extrusion of a blend of polymers which are incompatible such that on solidification the blend comprises a dispersion of one polymer in a polymeric matrix.

16. A method according to claim 1 substantially as herein described with reference to the drawings.

17. A laminate made by a method according to any preceding claim.

18. Apparatus for carrying out a method according to claim 1 comprising means for advancing two or more films towards a transverse stretching system, and means for superposing the films in the system, in which the transverse stretching system includes means for applying pressure along lines extending substantially in the longitudinal direction of the films and for simultaneously stretching the films in the transverse direction, thereby forming a laminate having a configuration of temporarily substantially evenly distributed substantially longitudinal pleats.

19. Apparatus according to claim 18 in which the means for applying pressure and for stretching the films comprises a pair of intermeshing grooved rollers and means for passing the films through the lip of these rollers, the grooves extending substantially in the longitudinal direction of the films.

20. Apparatus according to claim 19 comprising a plurality of pairs of the intermeshing grooved rollers and means for passing the films through the nip of each of these in series.

21. Apparatus according to claim 19 or claim 20 including means for heating at least one of the grooved rollers.

22. Apparatus according to any of claims 18 to 21 further comprising means for longitudinally stretching the laminate.

23. Apparatus according to claim 22 in which all means for longitudinally stretching the laminate are positioned for stretching the laminate after it has been transversely stretched.

24. Apparatus according to claim 18 substantially as herein described.

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1526724 COMPLETE SPECIFICATION

2 SHEETS *This drawing is a reproduction of the Original on a reduced scale*

Sheet 1

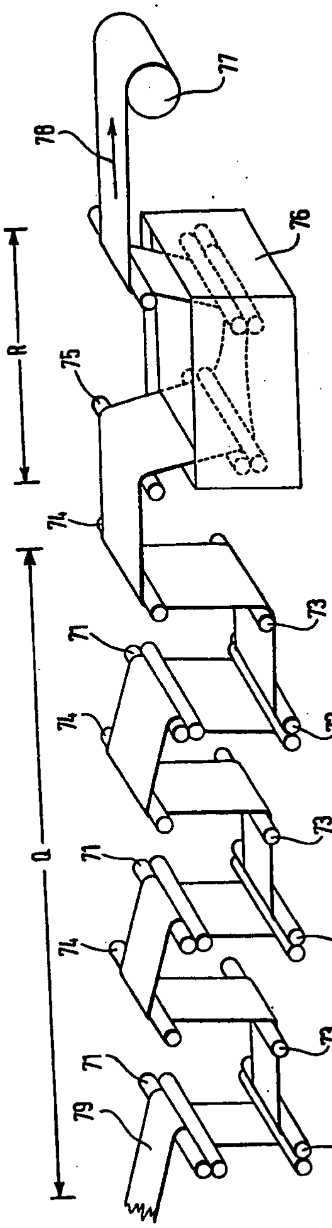

Fig. 1

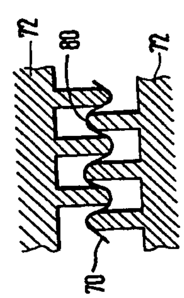

Fig. 2

Fig.3

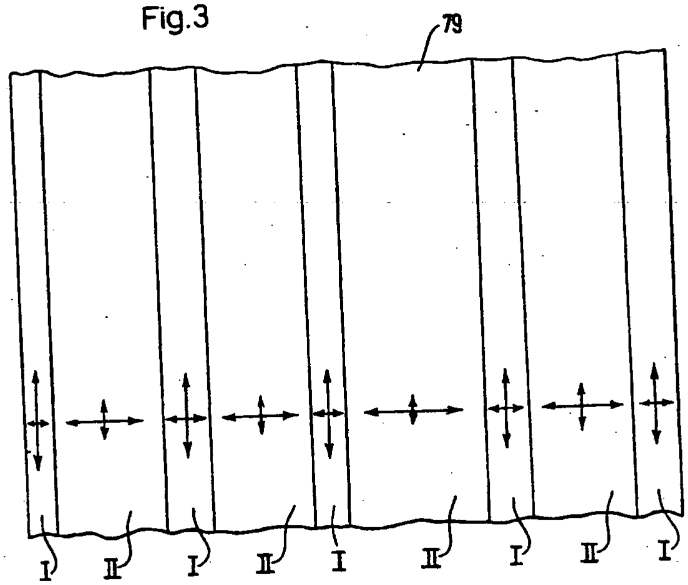

Fig. 4